

Cooper pairs without ‘glue’ in high- T_c superconductors

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Abstract –We address the origin of the Cooper pairs in high- T_c cuprates and the unique nature of the superconducting (SC) condensate. Itinerant holes in an antiferromagnetic background form pairs spontaneously, without any ‘glue’, defining a new quantum object the ‘pairon’. In the incoherent pseudogap phase, above T_c or within the vortex core, the pairon binding energies are distributed statistically, forming a ‘Cooper-pair glass’. Contrary to conventional SC, it is the mutual pair-pair interaction that is responsible for the condensation. We give a natural explanation for the *ergodic rigidity* of the excitation gap, being uniquely determined by the carrier concentration p and J . The phase diagram can be understood, without spin fluctuations, in terms of a single energy scale $\sim J$, the exchange energy at the metal-insulator transition.

Introduction.

A remarkable aspect of the BCS superconducting (SC) state is its universality, explaining a large number of properties quite independently of the detailed composition of the material [1]. The conventional SC state emerges from a general metallic state leading to a macroscopic wave function responsible for zero resistivity, perfect diamagnetism and Josephson quantum effects [2]. The microscopic picture is the condensation of Cooper pairs [3] wherein the binding energy is the order parameter, vanishing at T_c , and whose $T = 0$ value is material independent, with a universal ratio : $2\Delta/k_B T_c \simeq 3.52$. Although conventional SC is well understood, the pairing mechanism due to phonon exchange [4] is still hidden in the ground state energy gap. However it is revealed in the strong-coupling quasiparticle (QP) excitation spectrum in the fine structure at the phonon energies above the gap [5,6].

In marked contrast, high- T_c superconductivity [7] emerges upon doping from an insulating state dominated by antiferromagnetic (AF) interactions [8]. The macroscopic wavefunction expresses the hallmark SC properties, but the microscopic mechanism, in particular the pair for-

mation, remains unknown. Moreover, the relevant parameters are orders of magnitude different from the conventional case : the nanometric coherence length, a large spectral gap, large penetration length and, of course, very high T_c . Strikingly, the magnitude of the spectral excitation gap, as measured by tunneling [9,10] and photoelectron spectroscopies ([11] and refs. therein), remains constant as a function of temperature up to and just above T_c , in the pseudogap (PG) state (see [12] and refs. therein), suggesting the existence of pairing correlations above T_c [13,14]. Thus, contrary to the conventional BCS scenario, the energy gap is clearly not the order parameter.

In this work, we propose a simple mechanism for the pair formation in high- T_c cuprates. We demonstrate that hole pairs in a lightly-doped antiferromagnetic environment are energetically stable. Their binding energy is directly related to the exchange energy J of the surrounding electron spins. Thus, the hole pairing arises spontaneously, without ‘glue’, due to an effective quantum potential well. This defines a new complex quantum state, a hole pair coupled to its local AF environment, the ‘pairon’.

Beyond the AF phase, the pairons are in a disordered incoherent state, a ‘Cooper-pair glass’ wherein their binding

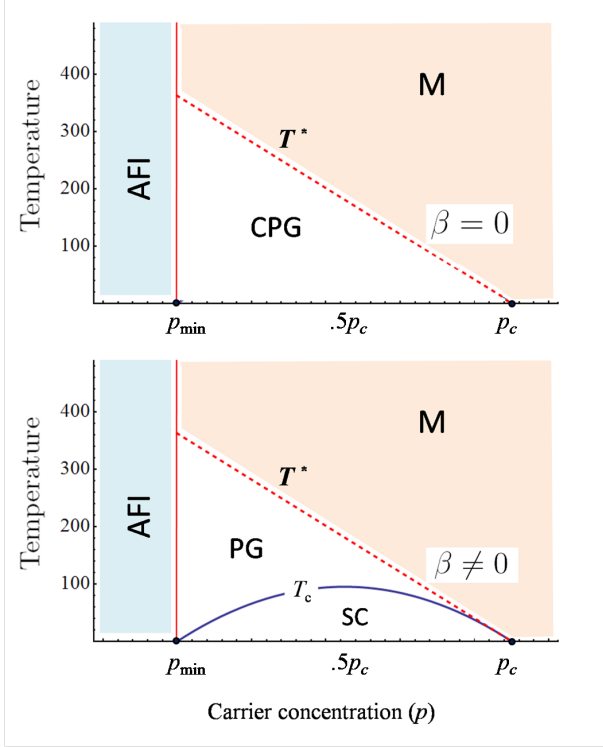


Fig. 1: Phase diagram as a function of doping without pair-pair interaction ($\beta = 0$, upper panel) and with PPI ($\beta \neq 0$, lower panel). As a result of the PPI the critical dome separates the PG phase from the SC phase at lower temperature.

energies are statistically distributed. This state is revealed above T_c in the pseudogap (PG) phase, and also within the vortex core once SC coherence is lost [15]. Contrary to the BCS theory, the SC coherent state is achieved due to the mutual pair-pair interaction β (PPI), as shown in the phase diagram, Fig. 1, lower panel.

A novel fundamental constraint emerges in this work: the condensate wave function is such that the hole-pair/antiferromagnetic state (pairon) must be homogeneous. As a result, the spectral gap is pinned to the specific value Δ_p which is remarkably constant with regards to perturbations such as temperature, magnetic field or disorder. We show that the latter gap value is uniquely determined by the doping p and the exchange energy J . This pairing constraint suggests the novel concept of ‘ergodic rigidity’.

Contrary to conventional SC, the pairing energy Δ_p is not the condensation energy. Rather, it is the mutual interaction between pairs that allows for a Bose-type condensation onto the ground state [16, 17]. Consequently, $\beta(T)$ is proportional to the condensate density which decreases with temperature due to the pairon excitations, instead of quasiparticles, to finally vanish at T_c . The PPI has the required properties of an order parameter and in this work we show that it is a unique function of p and J .

The SC state is thus built from two inter-dependent phenomena, the pairon formation and their mutual interaction, both depending on a single energy scale J .

Origin of Cooper pairs in cuprates

In high- T_c cuprates, superconductivity emerges upon doping from the antiferromagnetic state at half filling i.e. one electron per copper site. As a function of doping, it starts at a minimal value $p_{min} \approx 0.05$, corresponding to the extinction of the long-range magnetic order ($T_{N\acute{e}el} \sim 0$). In spite of the vanishing of the AF order parameter, antiferromagnetism is known to exist at the local scale, as indicated by the paramagnon resonance even inside the SC dome [18]. A large number of measurements have confirmed the small SC coherence length of the Cooper pairs in these materials ($\xi_{pair} \sim 1 - 2 nm$). Thus, provided that the magnetic coherence length ξ_{AF} is large compared to the lattice constant, the exchange energy J is logically the dominant energy scale.

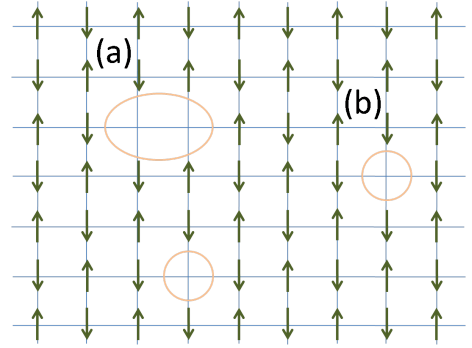


Fig. 2: Representation of two separate holes as well as one hole pair in an AF background. Due to the magnetic energy gain, the hole pair configuration is favorable compared to two separate holes.

In this AF background, pairs of holes are energetically more favorable than individual holes (see Fig. 2). Considering only the magnetic potential energy for the square lattice,

$$H_{AF} = J \sum_{i \neq j} \vec{S}_i \cdot \vec{S}_j$$

where i and j are nearest neighbor occupied sites. Thus, two individual holes have an energy $8J$ with respect to the full AF ‘sea’, while a hole pair has an energy $6J$, leading to a gain of J per hole. Thus, in the nearest-neighbor approximation, the energy gain for a single hole pair in a perfect infinite AF background is J compared to two separate holes. This provides a simple explanation for the pairing in these materials – they arise spontaneously without ‘glue’, i.e. *without a boson exchange*, due to the surrounding AF background.

This effect can be viewed as a quantum potential well of depth J for two holes. The question then arises whether 4 holes together, or a more numerous populated island, would not be more stable. In fact they are not since the interaction between pairs is repulsive. Indeed, the effective potential well becomes rapidly nil when two pairs are

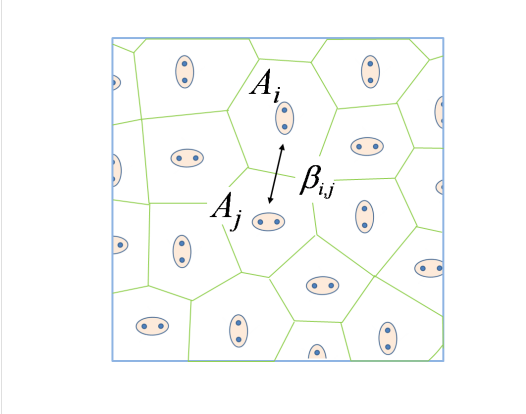


Fig. 3: Snapshot spatial representation of the Cooper pair glass. Each pair is associated with an AF area \mathcal{A}_i which determines the binding energy Δ_i of the pairon.

approached together. Given these considerations, the expected stable solution for itinerant holes in an AF background should be preformed pairs.

We now consider a statistical ensemble of uncondensed pairs of density $p/2$, each pair being surrounded by a number of nearest neighbors as in Fig. 3. Clearly, the binding energy for each pair must be directly affected by the local AF environment. This conclusion is in good agreement with the experimental AF correlation length as function of doping [19]. Remarkably, $\xi_{AF} \sim 1/\sqrt{p}$, which is also the typical distance between pairs.

The simplest model, which works remarkably well, is that each binding energy is uniquely determined by the area \mathcal{A}_i available to each pair. This defines a new composite object, *the pairon*, being a quantum state consisting of a hole pair entangled with its local AF environment. Thus, we take the i th pairon binding energy Δ_i to be proportional to the corresponding area \mathcal{A}_i which, in a classical view, is the Voronoi cell area in a two-dimensional distribution [20].

For large density, an additional consideration is necessary. As p increases, and the characteristic areas decrease, there is a minimum incompressible area \mathcal{A}_c at which the local AF environment is too small to create a potential well. In this limit, the binding energy for a pairon vanishes. The minimum configuration is one hole pair surrounded by 6 occupied copper sites, giving a critical doping of $p_c = 2/8 \approx 0.25$. This agrees well with the observed critical value for $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ ($p_c \simeq 0.27$). This effect is also related to the critical percolation threshold for holes on a square lattice corresponding to 4 sites for each hole [21].

For small densities, the critical point for SC onset is p_{min} ($p_{min} \simeq .05$ for $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$). This value corresponds to the largest Voronoi cell area wherein the pairs can condense. At this point, their binding energies are in fact maximum with a binding energy close to J .

With these considerations, between these two limits, the

pairing energy for each pair i is given by :

$$\Delta_i = J_{eff} (\mathcal{A}_i - \mathcal{A}_c) / \langle \mathcal{A} \rangle \quad (1)$$

where J_{eff} is the effective exchange energy, \mathcal{A}_i is the Voronoi area for the i th pair (see Fig. 3) and $\langle \rangle$ denotes the average. As a result, the mean binding energy is :

$$\Delta_0 = \langle \Delta_i \rangle = J_{eff} \times (1 - \frac{p}{p_c}) \quad (2)$$

i.e. simply determined by the number of holes and J_{eff} . This general relation shows that the dominant effect is the linear decrease of the pair binding energy with the carrier concentration to ultimately vanish at the critical point p_c , a universal feature of high- T_c superconductivity.

We now consider the many-body properties of the pairon system.

The Cooper-pair glass state

Unconventional superconductivity in cuprates can be described by a microscopic Hamiltonian of interacting preformed pairs [16]. In absence of pair-pair interactions, the scenario is an incoherent state of preformed pairs that are distributed in different energy states defined by their binding energies Δ_i . We call this state the Cooper-pair glass (CPG), illustrated in Fig. 3. It is quite analogous to the Bragg-glass vortex phase [22, 23], unique to high- T_c superconductivity, which is formed when the vortex-vortex interaction (the PPI in the present case) is small compared to any local disorder potential.

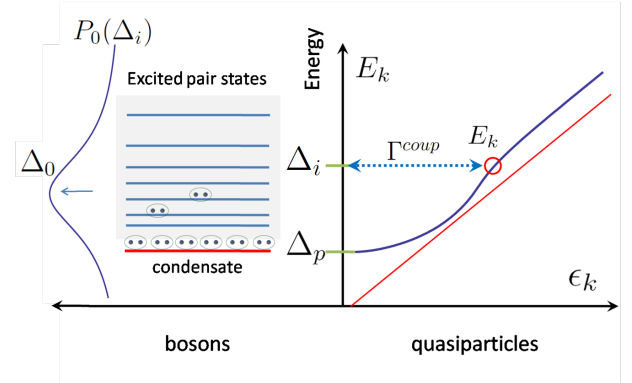


Fig. 4: Boson/pair degrees of freedom (left side) and fermion/quasiparticle excitations (right side). The degeneracy between quasiparticle energy E_k and the excited states energy Δ_i , leads to the dip singularity in the QP DOS.

To understand the CPG state, characterized by Δ_i , we note that the CPG Hamiltonian can be formally mapped onto the Suhl-Mathias-Walker model for multiband superconductors [24] where i is the band index. In our case, the i th index refers to the given CPG pair state. The corresponding density of states is very well described by a Lorentzian form :

$$P_0(\Delta_i) \propto \frac{\sigma_0^2}{(\Delta_i - \Delta_0)^2 + \sigma_0^2} \quad (3)$$

where Δ_0 and $2\sigma_0$ are respectively the median and the width of the pair energy distribution (see Fig. 4, left panel). Again, $P_0(\Delta_i)$ reflects the variation of the possible configurations of the Voronoi cells discussed previously.

Superconducting coherence arises as a result of the mutual interactions between pairons, leading to a Bose-Einstein type condensation into the single energy state $\Delta_i = \Delta_p$. From the microscopic Hamiltonian [16], the latter must satisfy the self-consistent gap equation :

$$\Delta_p = \Delta_0 - 2\beta^c P_0(\Delta_p) \quad (4)$$

where β^c is the interaction energy in the SC state described below. In the absence of the PPI, i.e. $\beta^c = 0$, the pairons are distributed in the energy states centered around Δ_0 , the Cooper-glass state. The excitation spectrum exhibits a broad gap without quasiparticle peaks (Fig. 5), as observed in tunneling experiments when coherence is broken, either at the critical temperature [9, 10, 25] or locally within the vortex core [15].

Below T^* the CPG state is more favorable than the normal metallic state but, due to pair-pair interactions, it is unstable to SC condensation giving rise to the well-known critical dome (Fig. 1, lower panel).

The unconventional superconducting state

The accurate temperature dependence of the quasiparticle spectra measured by Sekine et al. [10] leads to the counter-intuitive conclusion that the spectral gap value, Δ_p , remains constant right up to the critical temperature. Significantly, this unique gap value is very robust as a function of external perturbations, temperature, magnetic field and disorder. Even though there are thermal pair excitations, so long as the condensate is non vanishing, the gap value is pinned to Δ_p . Since the system remains statistically invariant in spite of the perturbation, this leads to the novel concept of *ergodic rigidity* specific to unconventional SC.

Therefore, we take as a principle that in the SC state, for which $\beta^c \neq 0$, the gap value is imposed by the fundamental properties of the system, the doping p and the effective exchange energy J_{eff} . The coherence in the SC phase imposes homogeneity, which we obtain in the Voronoi scheme (equation 1) by choosing all areas \mathcal{A}_i to be equal. Thus, $\mathcal{A}_i = \mathcal{A}_{tot}/N_p$, where N_p is the total number of pairs and \mathcal{A}_{tot} the total area, guaranteeing homogeneity. In addition, we assume that in the uniform SC state $J_{eff} \rightarrow J$, where J is the exchange energy. With these considerations, we have :

$$\Delta_p = J \times \left(1 - \frac{p}{p_c}\right) \quad (5)$$

as indicated in table 1. In the SC state the spectral gap is thus uniquely determined by p and J . Since in the SC state the PPI is positive ($\beta^c > 0$), comparing the gap equation (4) with equation (2) leads to $\Delta_0 > \Delta_p$ and thus $J_{eff} > J$.

As a consequence of ergodic rigidity, the gap amplitude cannot be the order parameter of the SC to PG transition. As opposed to conventional SC, at finite temperature the system is not characterized by quasiparticle excitations (pair breaking) but by *pair excitations* following Bose-Einstein statistics [16], leading to an unconventional shape of the measured specific heat [26].

Superconducting coherence is subtly hidden in the ground state Δ_p but, contrary to BCS, it is not directly linked to the superfluid density. Indeed, examining the gap equation (4), the SC order parameter is the PPI term, $\beta^c(T) \propto N_{oc}(T)$, the latter being the number of condensed pairs. It should be measurable in any experiment sensitive to SC coherence such as Josephson effects [2]. In quasiparticle tunneling, the spectral gap remains constant, but the coherence peaks decrease monotonically due to the increasing pairon excited states leading to the pseudogap at T_c [17].

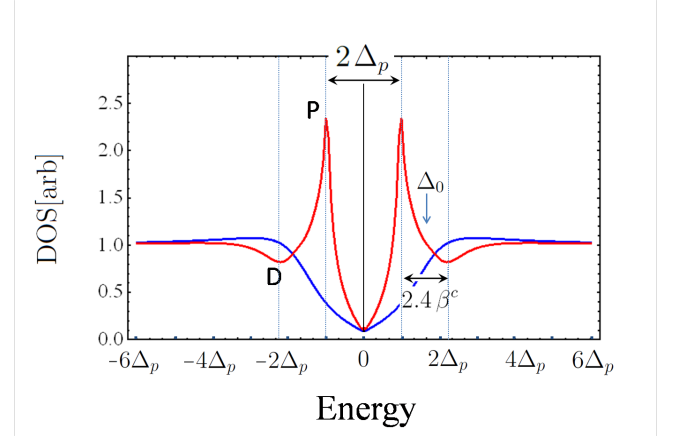


Fig. 5: Quasiparticle excitation spectrum calculated within our model in the Cooper glass state ($\beta^c = 0$) and in the superconducting state ($\beta^c \neq 0$) using equation (6) near optimal doping. Note that the shape of the coherence peak and dip structure in the SC state accurately fits the experimental tunneling data [16].

The PPI energy is universally revealed in the shape of the quasiparticle excitation spectrum beyond the gap energy measured by tunneling and ARPES (see [27] and refs. therein). This well known ‘peak-dip-hump’ fine structure is usually interpreted in terms of a spin-collective mode [28]. However, this compelling interpretation fails to fit the shape of the experimental spectra and does not account for the pseudogap phase.

The resolution of the microscopic Hamiltonian [16, 29] leads to the alternative conclusion that, due to the PPI, the quasiparticles are now coupled to excited pairon states (see Fig. 4, right panel). These ‘super-quasiparticles’ are described by an unusual dependence of the gap upon quasiparticle energy E_k :

$$\Delta_k(E_k) = \Delta_{0,k} - 2\beta_k P_0(E_k) \quad (6)$$

where the anti-nodal direction is assumed. The corre-

SC parameters

spectral gap	Δ_p		$J(1 - p/p_c)$
pair-pair int.	$1.2\beta^c$		$J(p/p_c)(1 - p/p_c)$
dist. maximum	Δ_0	$\Delta_p + 1.2\beta^c$	$J(1 - p^2/p_c^2)$
dist. width	σ_0	$\Delta_0/2$	$J(1 - p^2/p_c^2)/2$
dip energy	E_{dip}	$\Delta_p + 2.4\beta^c$	$J(1 + 2p/p_c)(1 - p/p_c)$

Table 1: Universal dependence of the parameters with doping. All parameters depend on the unique energy J . The doping p and the exchange energy J are measured from the SC onset (p_{min}). These equations are plotted in Fig. 6

sponding DOS (see Fig. 5) reveals the strong ‘peak-dip’ structure in the excitation spectrum seen in both cuprates [27] and iron-based superconductors [30, 31]. Using the parameters of Table 1, the theoretical curves fit accurately the experimental spectra for both classes of materials [29]. The peak-dip structure present at low temperature is thus a clear signature of long-range SC coherence, while the mere presence of a spectral gap is not.

We thus see clear deviations in the unconventional case from BCS in all aspects, be it the SC ground state, the pair (boson) excitations or the quasiparticle (fermion) excitations.

Universal dependence of the parameters on J

How does the pair-pair interaction depend on the fundamental parameters? In the mean-field treatment of the microscopic Hamiltonian leading to equation (4) the PPI is repulsive and can be expressed as: $\beta_c \propto p \times \Delta_p(p)$. From the detailed fits to the phase diagram (Fig. 6, inset) the proportionality constant is found:

$$\beta_c = 0,83 \times J \left(\frac{p}{p_c} \right) \left(1 - \frac{p}{p_c} \right) \quad (7)$$

where both J and p are measured from the SC onset.

This form of the mutual pair interaction can be visualized as the lowering of the potential well due to the interaction with neighboring pairs. Most significantly, both the gap magnitude Δ_p and the interaction energy β_c depend on a single characteristic energy J . This shows unambiguously that the pair formation (pairon) as well as the PPI depend on the same microscopic mechanism, the intimate cooperation of holes with the underlying antiferromagnetism.

As a function of the carrier concentration p , both Δ_p and β^c display a *universal dependence* upon doping (Fig. 6). The binding energy Δ_p is given by the exchange energy J at low doping and decreases linearly with p , while the PPI exhibits the universal dome, equation (7), as expected for the SC order parameter.

As shown in Fig. 6, all the parameters are simply proportional to the exchange interaction energy, with typical value $J \sim 100 - 130 \text{ meV}$ at half-filling in this class of materials [32]. Since the critical temperature scales with β^c ($\beta^c \simeq 2 k_B T_c$ for $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$) this energy scale explains the high critical temperature in cuprates.

Examining the gap equation (4) we note that Δ_0 , the average gap in the CPG state, is approximately given by: $\Delta_0 \simeq \Delta_p + 1.2\beta^c$ leading to the effective exchange energy $J_{eff}(p) \simeq J(1 + 1.2p/p_c)$. Thus as function of p , $\Delta_0(p)$ is a monotonic concave curve for the whole doping range. In addition, as indicated in Table 1, the distribution width $2\sigma_0$ is very close to Δ_0 , which is verified empirically (see inset in Fig. 6). Note that $\Delta_0(p) - \Delta_p(p) \simeq 1.2\beta^c(p)$ defines the *condensation energy* for any value of p .

As mentioned previously, the PPI is revealed in the dip position E_{dip} in the quasiparticle spectrum which has a doping dependence also shown in Fig. 6. It lies above $\Delta_0(p)$ for the whole doping range and is such that $E_{dip} \simeq \Delta_0(p) + 1.2\beta^c$. The latter relation agrees very well with experiment [16].

Many authors invoke the quasiparticle strong coupling to the spin collective mode [28] to account for the dip energy (relative to the gap Δ_p) which is found to be $\sim 4.9 k_B T_c$. In our work, the PPI is $\beta^c \simeq 1.8 k_B T_c$ which gives $E_{dip} - \Delta_p$ to be $4.4 k_B T_c$, in good agreement. As mentioned, our interpretation is completely different: The dip position is a direct consequence of the static quasiparticle-pair interaction expressed in Eq. (6).

The apparent contradiction can be lifted by noting that both depend on the exchange energy J . In particular, at optimal doping our model indicates the precise value $E_{dip} = J$. Since both phenomena, the superconducting state and the paramagnon resonance, depend on the same energy parameter they are indeed highly correlated. On the other hand, as is well known in diverse fields, even a strong correlation does not imply causality.

The interpretation of the phase diagram is thus as follows: the subtle interplay between hole doping and antiferromagnetism leads to a pairing, without boson exchange. Contrary to the BCS case where Cooper pairs arise from the metallic state [3], here pairs emerge from an AF state. A crucial point is that the hole pair state (pairon) preserves at best the symmetry of the AF lattice. Such a pairing mechanism would not exist in a frustrated spin system, such as in a triangular lattice, since in this case there is no energy gain when forming hole pairs. This prediction is in agreement with the absence of a SC transition in a Kagome-lattice spin liquid [33].

The pairons in this scenario cannot condense without the PPI and would remain in the Cooper glass state. How-

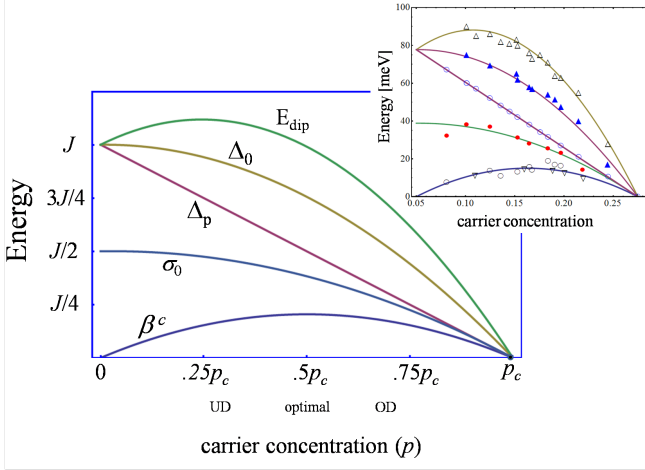


Fig. 6: Universal doping dependence of the parameters (see Table 1): The gap magnitude Δ_p , the pair-pair interaction β , the excited state parameters Δ_0 and σ_0 , and the dip position E_{dip} . The variation is shown from $p = 0$ at the SC to p_c the critical doping. Note the remarkable values: At optimal doping, β^c is maximum while the dip position is at $2\Delta_p$ such that $E_{dip} = J$. The maximum value of E_{dip} is at $p_c/4$. Inset: detailed fit for $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ using the equations of Table (1).

ever their mutual interaction, itself mediated by the same underlying AF local order, allows for the condensation onto the homogenous and unique state with energy Δ_p . As a consequence, the mutual interaction energy, β^c , as well as the pair formation mechanism, depend on the unique energy J .

How does this picture translate to the quantum mechanical formulation? Each hole pair in the AF sea, such as in Fig. 2, is surrounded by equivalent degenerate pair positions where it can easily tunnel with no effect on the surrounding AF symmetry. Its wave function is thus:

$$|\Psi_{\text{pairon}}\rangle = \sum_{i,j} \alpha_{i,j} \hat{c}_{i\downarrow} \hat{c}_{j\uparrow} |AF\rangle \quad (8)$$

where i and j are nearest-neighbours and the Fermi operator $\hat{c}_{i\downarrow}$ creates a hole in the AF half-filled state $|AF\rangle$ at the site i . These pairon states are highly favorable and, when interacting with neighboring pairons, i.e. the PPI process, allows for a macroscopic SC wave function to be established.

Conclusion

In summary, in this article we show that the microscopic origin of high- T_c superconductivity is a subtle co-operation of holes with the underlying local antiferromagnetism. First, preformed hole pairs, the pairons, arise without ‘glue’ in the AF background with a binding energy directly related to the exchange AF energy J . The superconducting phase emerges from an incoherent disordered pair state, a Cooper-pair glass. As a result of pair-pair interactions, they condense following the Bose-

Einstein statistics onto a single pair state. The pair binding energy Δ_p is anchored to a value determined by the basic properties of the system, the exchange energy J and the doping p . The latter is remarkably robust with respect to a perturbation, be it temperature or magnetic field, suggesting the novel concept of *ergodic rigidity* at the heart of the transition.

Contrary to BCS, the order parameter is the pair-pair interaction energy β^c . The unconventional SC state is the subtle compromise between pairon formation, of high binding energy, and their repulsive interaction, both determined by the unique energy J . The high- T_c of cuprates thus follows. The single parameter J accounts quantitatively for the experimental phase diagram in (T, p) plane, the quasiparticle density of states and its evolution as a function of temperature in both the superconducting and the pseudogap phases.

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